Elicitation of dissolution rate data for potential wasteform types for plutonium under repository conditions

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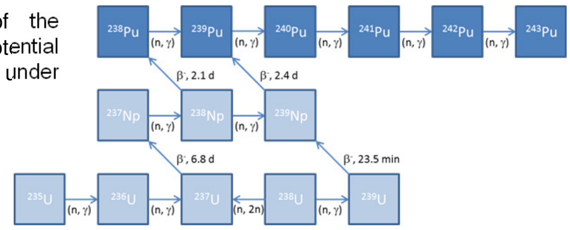
Background: In the UK, plutonium generated in uranium-based fuels during the operation of nuclear reactors is recovered during spent fuel reprocessing and stored mainly as PuO_2. The current preferred policy in the UK regarding the long-term management of separated civil plutonium is reuse as MOX fuel[1]. However, at least a fraction of the UK plutonium inventory is likely to be destined for geological disposal.

Objectives: Review and evaluation of the performance and long-term behaviour of potential wasteforms for plutonium disposal under repository conditions relevant for the UK.

Focal points
- durability in aqueous environments
- stability over geological time scales
- criticality control



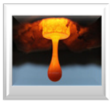
Civil plutonium

Source
- generated in uranium-based nuclear fuels by neutron capture
- separated during spent fuel reprocessing
- stored mainly as calcined PuO_2

Stocks of separated civilian plutonium in 2010[2-4]
- UK *c.* 92 t_{HM} (as of 31/12/2012)
- France *c.* 56 t_{HM}
- USA *c.* 54 t_{HM}
- Russia *c.* 48 t_{HM}
- Japan *c.* 45 t_{HM}
- Germany *c.* 12 t_{HM}

High-level disposition options for separated plutonium
- ➢ long-term storage (and disposal)
- ➢ recycling/reuse (MOX fuel; inert matrix fuel)
- ➢ immobilisation and geological disposal

Potential plutonium wasteforms



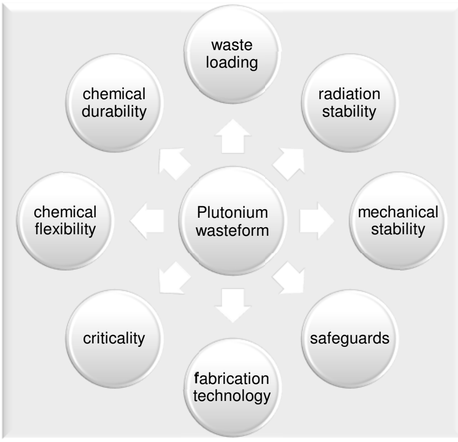
- Glasses
  - Borosilicate glasses
  - Alkali-Tin-Silicate glasses
  - Phosphate glasses

- Ceramic wasteforms
  - polyphase (Synroc)
  - single-phase (e.g. pyrochlore, zircon, zirconolite, monazite, …)

- "storage" MOX
  - MOX fuel not destined for reactor usage (fabricated by established technology with reduced technological specification)

- Encapsulants
  - Cement
  - Polymers

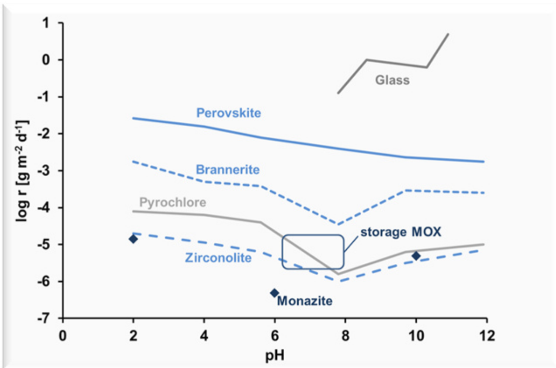
Relevant issues for plutonium wasteforms



Review of wasteform performance

- Knowledge base on plutonium wasteform behaviour highly variable focused mainly on ceramics and glasses.
- Investigations into plutonium wasteform durability and radionuclide leaching are to a large extent performed with (short-term) static test methods (e.g. MCC, PCT) often using deionised water.
- Only few data on wasteform dissolution behaviour obtained in long-term tests or under dynamic conditions in flow-through or column tests.
- Surrogates such as cerium, hafnium or neodymium used in most investigations instead of plutonium.
- Corrosion/leaching rates of ceramic wasteforms such as pyrochlore, zirconolite, or monazite (and storage MOX) generally significantly lower compared to glasses.
- Limited direct applicability of leaching test data to repository conditions and for model development.
- Detailed understanding of relevant processes that govern wasteform corrosion, radionuclide release and total systems behaviour seems to be still missing.
- ➢ Derivation of "bounding values" for corrosion rates of generic wasteform types under disposal conditions from experimental data and analogue evidence.

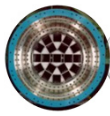
Selected experimental data for dissolution rates of potential plutonium wasteforms[5-8]



Concepts for geological disposal in the UK

Higher strength crystalline rocks
- granite, gneiss
- SKB KBS-3V concept (copper canister, bentonite buffer)

Lower strength sedimentary rocks
- clay, clay rock, mudrock
- NAGRA Opalinus Clay HLW/SF-concept (steel canister, bentonite buffer)

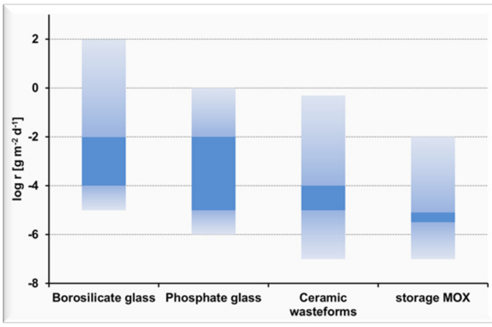


Evaporites
- bedded rock salt, anhydrite
- DBE drift concept (steel canister, crushed salt backfill)

→ Geological disposal programme in generic stage:
- no decision on host rock lithology
- illustrative disposal concepts (incl. ILW co-disposal)

→ wide range of possible near field conditions (e.g. pH 7 … 13; low/high salinity, …)

Derived Pu wasteform durability under disposal conditions



→ Expected ranges (dark blue) and bounding values for the corrosion rates of generic plutonium wasteforms under repository conditions derived from experimental data and analogue evidence (e.g. from HLW-glasses and spent fuel corrosion) using geochemical reasoning

Conclusions & Outlook

- Information on the long-term behaviour and durability of plutonium wasteforms under disposal conditions is rather limited compared to HLW-glasses and spent fuel.
- Bounding values for plutonium wasteform corrosion rates under repository conditions can be derived from available experimental data and analogue evidence.
- More realistic assessments of wasteform durability and radionuclide release behaviour would require systematic studies regarding
  - wasteform corrosion under realistic conditions,
  - secondary phases formed during wasteform corrosion, and
  - effects of self-irradiation on wasteform durability

  to explore the safety margins of the various potential wasteforms.

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Acknowledgement: This work was funded by the UK Nuclear Decommissioning Authority – Radioactive Waste Management Directorate (NDA RWMD)

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